Glyphosate and AMPA concentrations in wind-blown material under field conditions.

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ABSTRACT

Agricultural intensification in fragile arid and semi-arid environments has led to an increase in soil degradation, mainly through wind erosion. Argentina is an agricultural and cattle-farming country, which has increased its productivity in the last few decades, widening the boundaries of farm land and the use of herbicides to control weeds. Glyphosate, which accounts for 65% of the Argentinian pesticides market, is strongly retained in soil. The World Health Organization concluded that there was evidence to classify glyphosate as 'probably carcinogenic to humans'. In this context, the objective of this study was to determine the presence and concentration of glyphosate and aminomethylphosphonic acid (AMPA) in wind-blown material in three areas in Argentine semi-arid regions (Chaco, CH; La Pampa, LP and San Luis, SL). In one hectare (ha) squared plots, left uncovered and levelled, the wind-blown material was collected at heights of 13.5, 50 and 150 cm during 18 erosion events. The wind-blown material carried by the wind at a height of 150 cm had concentrations of 247µg kg^{-1} and 218µg kg^{-1} of glyphosate and AMPA, respectively.This material was enriched 60 times in glyphosate and 3 times in AMPA as compared to the original soil. This shows that the eroded material can, potentially, have a negative impact on the ecosystem and also on human health, depending on the proportion of this material released into the atmosphere in suspension as particulate matter. This study is, to our knowledge, the first to report concentrations of glyphosate and AMPA in wind-blown material under field conditions.

INTRODUCTION

In the last decades, there was an expansion of agricultural production on natural systems 'not intervened or scarcely intervened by man' as one of the strategies to increase production. These natural systems, fragile for agricultural production, suffer an important pressure due to their intensification (Thiombiano, 2000). This agricultural intensification has been seen in arid and semi-arid environments in the clearing and ploughing of soils that are highly susceptible to erosion by wind and rain, endangering the sustainability of the soil. Wind erosion is one of the most significant soil degradation processes in dry ecosystems (Colazo and Buschiazzo, 2015; Gao et al., 2015; Houyou et al., 2014; Peterson, 2006; Buschiazzo et al., 1999) and affects processes such as desertification, human health and climate change (Wang et al., 2015; Xie et al., 2015; Nicholson, 2000; Pope et al., 1995; U.S. EPA, 1995; Pope and Dockery, 1998). During wind erosion, soil is moved via three mechanisms known as creep (particles larger than 500 µm de diameter), saltation (particles between 100 µm and 500 µm) and suspension (particles less than 100 µm) (Chepil, 1945).The 90% eroded material moves by saltation at a height less than 30 cm and only 1% at over 1 meter high (Chepil and Woodruff, 1963). Saltation is important because it triggers the erosion process and increases the erosive effects in the prevailing wind direction, since as particles hit the ground, new particles disaggregate; aggregates are broken, themselves producing more eroded material (Chepil, 1957). Particles moving by saltation are transported from a few meters up to several hundred meters. Finally, particles moving by suspension can be carried hundreds or thousands of kilometers from the source (Füllen and Mitchell, 1991, Alfaro et al., 2004). The size of the particles carried by the wind decreases exponentially with height (Li, et al., 2008).

The material transported by saltation has the potential to affect soils, with decrease in the thickness of the surface layer, changes in texture, breakdown of structure and a loss of chemical fertility through the decrease of organic matter and nutrients (Aimar 2016, Lyles and Tatarco, 1986; Buchiazzo and Taylor, 1993; Gabel, 1993). The material transported by suspension somewhat has negative effects on the environment (by modification of marine trophic chains and change in the formation of clouds and radiation) and human health (because particles smaller than 10 microns can be lodged in the lungs and cause disease) (Grantz et al., 1998, Norton and Gunter, 1999, West et al., 2016).

Argentina has 220 million ha of land in semi-arid and arid climates (Fryrear, 1990), of which approximately 30 million ha are affected by wind erosion (PROSA, 1988). Previous studies have shown that wind erosion in the semi-arid central Pampa region causes losses of 5 to 1850 kg ha^{-1} for winter crops and 0 to 17500 kg ha^{-1} in summer crop (Mendez and Buschiazzo, 2010; Mendez and Buschiazzo, 2014). These studies also showed that even in direct sowing, wind erosion can exceed the allowed thresholds of 700 kg ha^{-1} in both summer and winter crops.

Moreover, grain production increased by 185.3% between the 1970-71 and 2012-13 growing seasons, causing an expansion of agricultural land into areas of greater environmental fragility (Viglizzo, 2011). In addition, the amount of area under cereal and oilseed cultivation in the 1970-71 growing seasons was 64.8% and 8.5%, respectively; after four decades, in the 2012-13 seasons, this amount had changed to 29.1%and 60.3%, respectively (SIIA, 2014).

The cultivation of soybean accounts for a significant part of agricultural land in the country, mainly due to the adoption of a technological package that involves the no-tillage system and the use of genetically modified glyphosate-resistant seed. There

are currently other genetically modified crops, such as maize and cotton, that are resistant to glyphosate. Herbicides account for 65% of the Argentine pesticides market and glyphosate is the top-selling herbicide (CASAFE, 2012).

Glyphosate can be degraded in the soil by microbiological activity (Rampoldi et al., 2014; Zablotowicz et al., 2009; Gimseng et al., 2009; Dick and Quinn, 1995). Desorption and degradation are processes that probably occur simultaneously (Zablotowicz et al., 2009). But this herbicide has too a strong affinity to soil particles, which contributes to its build-up in that matrix, mainly when it is sorbed by soil colloids. Some soil substances that can sorbe glyphosate are clays, phosphates, organic matter, aluminum oxides and iron oxides (Borggaard and Gimsing, 2008; Gimsing et al., 2004).It has been shown that the half-life times of glyphosate in the soil ranges from few days to 1000 (Borggaard and Gimsing, 2008, Bento et al., 2016). The main metabolite is aminomethylphosphonic acid (AMPA) and this molecule generally forms more rapidly than it degrades, which contributes to its accumulation in the soil (Simonsen and Fomsgaard, 2008). Aparicio et al. (2013) showed that glyphosate and AMPA are present in soil that is being farmed.It was also shown that the presence of glyphosate and AMPA is relatively more frequent in the dust emitted from wind eroded agricultural soils and sediments (Bento et al., 2017) than in water. This is because the degradation of glyphosate in surface waters is very rapid and its practically irreversible sorption on the soils and sediments (Maqueda et al, 2017).

Chang, et al. (2011) suggested that AMPA will be introduced into the atmosphere only by soil erosion because it is formed in the soil after glyphosate application. Peruzzo et al. (2008) shown that in soils the temporal variation of glyphosate levels depended directly on the time of application and the rain events. However, a later

study showed that the glyphosate concentration in the soil is more conditioned by the soil management history than by the date of the last application of the herbicide (Aparicio et al, 2013). In this way, the molecules could be concentrated in the soil and transported over long distances.

Previous studies in the semi-arid region of Argentina have shown that the material transported at a higher altitude (by suspension) is more enriched in colloids than the material transported near the soil surface and the original soil (Aimar, 2012 and 2016). Therefore, the herbicide particles could be more concentrated at a higher height. However, there are no reported data on glyphosate and AMPA concentrations in soils of arid and semi-arid regions susceptible to wind erosion, where soil losses from soil erosion up to 270 Mg ha^{-1} $years^{-1}$ have been measured in the field (Aimar, 2002). There is even less information about glyphosate and AMPA concentrations in material transported by the wind via saltation and suspension. The World Health Organization concluded that there is evidence to classify glyphosate as 'probably carcinogenic to humans' (Group 2A) (WHO, 2015) and, the experts on pesticide residues in food and the environment at a meeting of the FAO, concluded that glyphosate together with AMPA should be considered as residues of toxicological interest. For the purpose of estimating the dietary intake and to allow comparison of the calculated intakes with ADI, it is preferable to express the residues in terms of glyphosate (glyphosate = 1.5 × AMPA) (FAO report, 2005). For these reasons, it is important to assess whether the process of wind erosion contributes to glyphosate and AMPA redistribution in the environment.

This study aims to determine the presence and concentration of glyphosate and AMPA in wind-blown material in three areas of the Argentine semi-arid regions

(Chaco, CH; La Pampa, LP and San Luis, SL) that are in the agricultural production region, in soils with and without history spraying of glyphosate.

MATERIALS AND METHODS

Selection of experimental sites

Three experiments were conducted in semi-arid regions of Argentina, in fields located in the departments Almirante Brown (26°S 61°W, Chaco Province, CH), Capital (36°S 64°W, La Pampa Province, LP) and General Pedernera (33°S 65°W, San Luis Province, SL). The locations of the experimental sites in Argentina are shown in Figure 1. In CH and LP plains are mostly covered by soils classified as Mollisols and Entisols developed on Pleistocene and Holocene loess (Tricart, 1973). In SL the soils develop on Holocene Aeolian sands. Their texture is loamy-sand and sand. Dominant soils areTypic and UsticTorripsamments and Typic Ustisamments (Moscatelli and Puentes, 1996).

The sites were selected according to contrasting and the different agricultural history of each region. In CH site, plot was installed in a grass soil, which was ploughed to maintain the plot without vegetation and never application of herbicide was made. In the surrounding area there were patches with 2 and 3 years of annual crops (mainly soybean) and forest.The SL site has had five years of agricultural activity (mainly annual crops under no-tillage and a high use of pesticides). In the surrounding area, there were the same characteristics. The LP site has had four years of alfalfa pasture after four years of annual crops (oats, rye and sorghum) under conventional tillage with little use of pesticides. The surrounding area was similar management to the sampling plot. Since the installation of the erosion plots, weed control was always done mechanically.

Soil analysis and eroded material calculation

Soil samples composed of 5 sub-samples of 2 kg of weight were taken in each experimental site from a depth of 0-2 cm. Once air dried and sieved through a 2 mm sieve, the following analysis were performed: organic matter contents (OM) by means of the wet digestion method (Walkley and Black, 1934), grain size distribution by means of the Robinson-pipette method (Gee and Bauder, 1986) and aggregate size distribution by sieving with vertical rotary sieve. The sieves were superimposed vertically in a support that is mechanically activated. The soil sample was placed in the upper sieve, the engine was operated for 5 minutes. Thus the following aggregate sizes was separated: <50 µm, 50–75 µm, 75–100 µm, 100–250 µm, 250–840 µm and 840–2000 µm. The soils textures range from coarse to fine are SL>CH>LP and the organic matter content (OM) was in CH<SL<LP (Table 1).From the aggregate size distribution analysis the geometrical mean diameter of aggregates (GMDa) was obtained (Table 1). Concentrations of glyphosate and AMPA were also determined (Table 2).

Wind erosion sampling plots were established and eroded material was collected from November 2004 to November 2010. At each site, during the measurement period, all erosive dry events were quantified in CH (n= 48), LP (n= 34) and SL (n= 23). Were selected those erosive dry events where sufficient material was collected for analysis, CH (n = 7), LP (n = 8) and SL (n = 3). The plots were designed in one-hectare squares and the soil surface was left uncovered and levelled. The mechanical work was performed with a disc harrow to control weeds and to control crusting of the soil surface, whenever it was necessary. Four masts were placed in each plot, each one at the midpoint of each side of the plot (Figure 2). To quantify and collect eroded material in every dust storm were placed on each mast 3 BSNE (Big Spring Number Eight) samplers, designed by Fryrear (1986). These samplers

capture particles mobilized by saltation (mainly at heights of 13.5 and 50 cm) and, to a lesser extent, by suspension (mainly at a height of 150 cm), (Fryrear and Saleh, 1993) (Figure 2). During the sampling, there was no precipitation and there was no herbicide placement. Previous dates of herbicide application have not been taken into account because the concentration in the soil will depend on the previous management history (Aparicio et al, 2013) and the material evaluated in the collectors may come from other nearby plots. An automatic weather station was installed at the center of the plot to record the direction and wind speed every 5 minutes. Wind direction, maximum wind speed and storm duration were obtained from meteorological records. The storm duration is the number of minutes in which the wind speed exceeds the threshold speed, which is the minimum speed necessary for the process of wind erosion to start. The duration of the storm (DS) was calculated using the following equation:

$$DS = \sum_{i}^{n} WE_{\geq TV} \qquad \text{Eq. [1]}$$

where *DS* is the duration of the storm in minutes, i is the time during which the samplers were installed (in minutes), n is the time during which the samplers were removed (in minutes), *WE* is the average wind speed every 5 minutes and *TV* is the threshold speed (ms^{-1}). Erosive velocities were considered for those greater than 6.7 m sec^{-1} at 2 m, which is the threshold wind speed for the soils of central Argentina (de Oro and Buschiazzo, 2008).

In order to determine the magnitude of the erosive events, the horizontal mass transport (*HMT*) was calculated. This is the amount of soil passing by unit area of a horizontal plane defined between two definite heights. First calculate the horizontal mass flux (*HMF)*, is the amount of soil particles passing by unit area of a vertical

plane in each individual sampler, was calculated by dividing the amount of material by the size of the sampler opening (Eq. 2).

$$HMF = \frac{Q}{S} \qquad \text{Eq. [2]}$$

Where *HMF* (kg m^{-1}): is the horizontal mass flux, *Q* (kg): is the amount of soil caught by the BSNE sampler and *S* (m^2): is the inlet area of the BSNE.

The *HMT* was calculated by integrating an exponential function proposed by Stout and Zobeck (1996), which fits the horizontal mass flux (*HMF)* variation as a function of height, between 13.5 and 150 cm high (Eq. 3). The integrations were made between measured heights (of 13.5 and 150 cm), because small changes in the lower boundary for the vertical integration have different effects on the amount of material calculated with each equation (Funk et al., 2004; Panebianco et al., 2010).

$$HMF(z) = \sigma\, e^{-\beta / z} \qquad \text{Eq. [3]}$$

where, HMF (z) is the horizontal mass flux at the z height, z is the height and σ and β are just adjustment coefficients without any physical meaning.

Finally, the total amount of soil released from the plot was calculated by subtracting the HMT from the outlet edge of the samplers from the HMT from the inlet edge of the samplers on the plot. Prevailing wind direction during each wind storm was obtained from meteorological records. The sediments obtained were homogenized by height to extract pesticides in the laboratory.

Extraction and analysis of glyphosate and AMPA

A representative sub-sample of wind-blown material (2g) was overlaid with 15µL of isotope-labelled glyphosate (1,2-^{13}C, ^{15}N) stock solution (10 mg L^{-1}), taking care to

distribute it uniformly on the sediment, followed by a rest of 30 minutes to stabilize the system. After that, wind-blown material was extracted with 5 ml of $KH_2PO_4/Na_2B_4O_7$ buffer (0.1M, pH=9) in an ultrasonic bath for 30 minutes. It was then centrifuged at 3,500 rpm for 10 minutes and the glyphosate and AMPA in the supernatant (aliquot = 2 mL) were derivatized with 2 mL of fluorenyl methyl chloro formate (FMOC) reagent in acetonitrile (1 mg mL^{-1}). The tube was shaken vigorously and left overnight at room temperature (between 12 and 15 hours). After that, in order to eliminate the excess FMOC, a liquid—liquid extraction with 5 ml of CH_2Cl_2 and centrifugation at 3,000 rpm for 10 minutes was carried out. Finally, the aqueous phase was filtered through a 0.22 µm nylon filter and 20 µL of the final extract was injected into the UPLC-ESI-MS/MS system (Waters Milford, MA, USA).

For the chromatographic separation, an Acquity UPLC BEH C18 column (1.7 µm, 50 x 2.1mm) (Waters) fitted with an Acquity Van Guard BEH C18 pre-column (1.7 µm, 5 x 2.1mm) (Waters) was used. The flow rate for the mobile phase was 0.4 mL min^{-1}. The mobile phase was a time-programmed gradient using organic-free water modified with ammonium acetate 5 mM (phase A) and methanol modified with ammonium acetate 5 mM (phase B). The percentage of organic modifier (B) was changed linearly as follows: 0 min, 0%; 0.2 min, 0%; 2.5 min, 70%; 3.5 min, 100%; 4.5 min, 100%; 5.0 min, 0% and 6 min, 0%. The column was kept at 60°C and the sample manager was maintained at 8°C. The drying, as well as the nebulizing, gas was nitrogen, obtained from a nitrogen generator. The cone gas and desolvation gas flows were optimized at 2 L h^{-1} flow and 600 L h^{-1}, respectively. For operation in MS/MS mode, the collision gas was argon 99.995%, with a pressure of $4.04x10^{-3}$ mbar in the T-Wave cell. Positive ionization mode was performed using a capillary voltage of 3.0 kV. The desolvation gas temperature was set to 400°C and the source

temperature to 120°C. Dwell times of 0.10 s/scan were chosen. Masslynx NT v 4.1 (Waters) software was used to process quantitative data obtained from calibration standards and from samples.

Mean recoveries obtained for glyphosate and AMPA were 85% and 81%, respectively. Limits of detection were 0.36 µg kg^{-1} for glyphosate and 0.41 µg kg^{-1} for AMPA, whereas quantification limits were 1.19 µg kg^{-1} and 1.6 µg kg^{-1} for glyphosate and AMPA, respectively.

Enrichment rates calculations

The enrichment rates (ER) of glyphosate and AMPA were calculated for each sampling height, in the following way:

$$ER = M_{mc}/M_{so} \quad \text{Eq. [4]}$$

Where: M_{mc} is the concentration of the molecule (glyphosate or AMPA) of the material from the sampler and $M_{so}$is the concentration of the molecule in the soil.

Statistical analyses

Analyses of variance (ANOVA) were performed with Version 6.12 SAS software (SAS Institute, 2003). The data from different dates were analyzed as repeated measurements using a mixed linear model (PROC MIXED). The random effect was locations and the fixed effect was sampling height. Mean comparisons were evaluated with a significance level of 0.05 using LSMEANS.

RESULTS AND DISCUSSION

Glyphosate and AMPA concentrations in the soil

The glyphosate and AMPA concentrations in the first 2 cm of soil depth varied between sites as follows: in San Luis (SL)> La Pampa (LP)> Chaco (CH) (Table 2). Crop management, prior to the installation of the experiment, would explain glyphosate and AMPA concentrations in all cases. In SL there were annual crops and weeds were controlled with the use of glyphosate, which would explain the

higher concentrations. On the other hand, in LP, alfalfa was grown in the last four years, which explains the lower concentrations and in CH, although glyphosate was not used, very low levels were detected, possibly as a result of soil particles movement from surrounding plot where glyphosate was used to control weeds in soybean cultivation.

Glyphosate levels in the CH and LP soils, without and with little history of glyphosate application, respectively, were lower than reported by Aparicio et al. (2013) in agricultural soils in the south east of Buenos Aires in the Pampas (between 35 and 1502 µg $kg^{-1}$of glyphosate). Meanwhile, the soil of SL, with at least five years of genetically modified crops and glyphosate application, showed concentrations of glyphosate that were within the range of values found by Aparicio et al., (2013) in the soils of south-eastern Buenos Aires. Similar results were reported for AMPA, with lower levels in LP and CH, and within the range reported by the same authors (between 299 mg kg^{-1} and 2,256 µg kg^{-1}) in SL. These results show that soils in semi-arid regions receiving glyphosate can reach concentrations similar to those found in humid regions.

On the other hand, our results show that the AMPA concentration is greater than glyphosate concentration in all the areas (between 14% and 38%). Although AMPA also degrades in the soil, the speed of this process is significantly slower than for glyphosate, possibly because the adsorption of the particles is stronger. The higher concentration of AMPA, compared to glyphosate, can basically be explained by the fact that the half-life of glyphosate is less than that of AMPA (Bohm, et al., 2008).In Argentine soils, glyphosate half-life was in the range of 9 to 38 days; however, 90% of the dose was present in soil for between 58 and 390 days after application under controlled laboratory conditions (Okada et al., 2017). This result would indicate that

glyphosate is present in the soil for between 2 and 13 months, depending on the type of soil. During this period, successive applications of glyphosate are made, maintaining the possibility that more AMPA is generated by degradation and that it increases its proportion in the soil with respect to the glyphosate. Another study has also shown that the glyphosate and AMPA content's decreased from the surface up to 25 cm deep (Lupi et al. 2015) indicating that the highest concentration is found in the surface layer susceptible to be transported by the wind. However, the transport of this herbicide to the atmosphere is also produced by vaporization of droplets during local or surrounding applications, for this reason glyphosate may be present in stormwater runoff, with an average of 2.9 µg L-1 (Lamprey and Ruban, 2008). Due to their continuous introduction into the environment, glyphosate and AMPA can be considered as "pseudo-persistent" pollutants (Primost et al., 2017), which may be able to cause the same exposure potential as regulated persistent pollutants, since their high transformation and removal rates can be compensated for by their continuous input into the environment.

Glyphosate and AMPA concentrations in wind-blown material

There were statistically significant differences in the glyphosate and AMPA concentration between the locations ($p < 0.0001$), in favour of the SL site compared to the CH site and the LP site (Figures 3 and 3b). There were statistically significant differences in the glyphosate concentration by sampling height ($p<0,01$), in favour of the 150 cm height compared with the 50 cm or 13.5cm heights (Figures 4a and 4b). No interaction was detected between location and sampling height ($p<0.2$).

With respect to experimental sites, greater glyphosate and AMPA concentrations were detected in SL compared with CH and LP (Figures 3 and 3b). In SL the highest glyphosate- and AMPA contents were found at all heights, 216.5, 895.8 and 1298.5

µg of glyphosate by $kg^{-1}$of soil, 410.8, 1125.8 and 1426.4 µg AMPA by kg^{-1} of soil at 13.5, 50 and 150 cm in height, respectively. All those concentration were found in the second erosive event (SL2) sampled in SL whose main meteorological conditions are showed in table 3 .This result would be related to the fact that, in SL´s plot and its surrounding was used a land management typical of the humid region of Argentina, where approximately 5 kg of glyphosate $hectare^{-1}$ $year^{-1}$were used to produce genetically modified crops resistant to this herbicide (mainly soybean, corn and cotton). The increase in the area planted with GMOs was 560,780 hectares in the 2014-15 growing season compared to the 2000-01 growing season in SL. At the CH and LP sites, the increase was 324,650 and 372,600 hectares, respectively, for the same growing seasons (SIIA, 2015). Moreover, in the period January to December 2013, in Argentina it was found that 41% of all pesticides were used on fallow land, 36% on the soybean crop, 10% on maize and the remaining 13% on other crops (CASAFE, 2014). Most of the pesticides are applied in the fallow period, and is a practice that goes along with no-tillage, although is not exclusive to it.. Due to the uniformity of management in this site, the material possesses a greater amount of glyphosate regardless of the predominant wind direction.

In CH, where the plot is surrounded by forest or agricultural parcels, was verified the contribution of particles came from neighboring regions, for example in the material collected on 10/14/2009 (CH5, Table 3) where the glyphosate concentration at 150 cm (180.6µg kg^{-1}) was 100 times higher than that found at 13.5 cm of height and 50 times higher than that found at 50 cm of height (data not revealed). In CH the concentrations in the collected material will depend mainly on the predominant directions of the winds and the existence of input of material from other plots. In particles dissolved in water (material eroded by water from soils in crop production),

the maximum glyphosate and AMPA concentrations were 562.8 µg kg^{-1} and 210.4 µg kg^{-1}(Aparicio et al., 2013), and 548 µg $Kg^{-1}$and 475 µgKg^{-1}, respectively (Primost et al., 2017). In our study, the wind-blown material or eroded material transported by the wind has average concentrations of 348 µg kg^{-1} and 418 µg kg^{-1} for glyphosate and AMPA, respectively. The presence of these concentrations of glyphosate and AMPA in the eroded material, and the levels found in CH in an area where the soil is never sprayed with herbicide, is evidence of the use of glyphosate in areas surrounding the experimental sites or the migration of soil particles mobilized by the wind from the others areas.

Wind erosion is a natural phenomenon that depends on the wind speed and dynamic variables of the soil surface, such as mulch, roughness and moisture. Given this and the distance between sampling sites (over 500 km), the erosive storms were of different intensities and occurred on different days of the year. Table 3 shows the meteorological features of the erosion events sampled and the horizontal mass transport (HMT) eroded in each one. The greatest HMTs recorded were 2231.4 kg m^{-1} in LP, 1614.1 kg m^{-1} in CH and 883.2 kg m^{-1} in SL, the reading in the latter site was taken with storms with lower wind speeds. In SL, the higher concentration of glyphosate and AMPA in the material eroded by wind, means that the net transport is greater, meaning that there would be greater risk of redistribution of the herbicide over greater surface areas, even with storms with low wind speeds.

The glyphosate concentration in wind-blown material or eroded material increases with height (Figure 4a). The material transported by wind has a finer texture at higher heights (Zobeck and Fryrear, 1986a). Studies conducted in the arid and semiarid region of Argentina with BSNE collectors, showed that the material that transports to 13.5 cm in height prevails in particles with a size between 100 and 250 µm. At 50 cm

height, the two most abundant textural fractions are 20 to 50 and 100 to 250 μm in diameter, and at 150 cm in height, between 40 and 54% of the particles are <50 μm (Aimar, 2016). Thus, at 13.5 cm in height, mainly individual grains of sand with a low affinity to the glyphosate molecule. The distance travelled by these particles is a few meters, so it is assumed that the greater proportion of the material comes from the same plot sampling (Cooke et al., 1993). The height of 50 cm is the limit of the movement by saltation and suspension; these comprise individual grains of sand and aggregates of silt and clay. The distance travelled by these particles are several tens of meters to hundreds of meters. Therefore, the material collected at 50 and 150 cm comes from both the plot under study and the surrounding plots.

Glyphosate is thought to be immobile or slightly mobile in soil as it ionizes easily and, as anions, is adsorbed to the organic matter (OM) in soil, or can compete with phosphorus (P) in adsorption areas (Prata et al., 2003) and correlates with soil pH (Lupi et al 2015). It has been shown that the aggregates and/or particulates that are transported at a greater height are of a finer granulometry, have more capacity for adsorption and are generally found to be enriched with elements such as organic carbon (C) and essential nutrients (Zobeck and Fryrear, 1986b; Aimar et al., 2014). Therefore, the increase in glyphosate and AMPA with height would be connected with the presence of the herbicide in adsorbed form to organic and inorganic colloids transported to a higher height, where the eroded material is finer (Aimar, 2002 and 2016; Zobeck and Fryrear, 1986a).In the laboratory, it was found that glyphosate and AMPA contents are highest in sediment particles <10 μm (PM_{10}), and that their content diminishes with increasing particle size (Bento et al., 2017). The authors mention that the risk of off-site airborne transport of glyphosate and AMPA with dust is, therefore, very high.

Glyphosate accounts for 65% of the Argentine pesticide market. This indicates the need to conduct further studies in natural conditions by improving the quantification and the separation of PM_{10} particles in the eroded material transported to wind at different heights and quantify the levels of pesticides in them. If glyphosate and AMPA-contaminated PM_{10} fractions of soil are emitted to the atmosphere, they may be inhaled by humans and animals. Inhalation of PM_{10} enriched with glyphosate means the entry into our body of a molecule classified by the IARC as probably carcinogenic (WHO 2015).

Enrichment rate (ER)

There were statistically significant differences for the *ER* of glyphosate between experimental sites ($p<0.0006$) where *ER* CH>LP=SL (Figure6). The *ER* increased with increasing heights. In this way, the eroded material at 13.5 cm height in CH, had an *ER* of almost double than LP and SL, and of 7 and 15 times greater at 50 and 150 cm, respectively. In CH there were statistically significant differences between sampling heights ($p<0.0339$) in favour of 150 cm height compared with heights of 50 cm and 13.5 cm. There was interaction between location and height ($p>0.0112$).

There were statistically significant differences for the *ER* of AMPA between experimental sites ($p<.0001$) in favour of CH over SL and LP, but not between heights ($p= 0.0606$) (Figure 6). There was no interaction between location and height ($p=0.0648$).

The material collected at the CH site was enriched with glyphosate, at the three heights (Figure 5). The higher *ER* of CH to the collectors located to 150 cm is probably because the contribution of particles came from neigh boring regions with agricultural plots and there were more particles of between 2 and 50µm in diameter transported in suspension (Aimar, 2016) and because of the low concentrations of

both molecules in the soil because it had never been sprayed with the herbicide. There was also a greater enrichment in AMPA seen in material collected in CH (Fig. 6).

These preliminary results show high concentrations of glyphosate and AMPA in wind-blown material. More studies are needed concerning the concentration of glyphosate and AMPA in different sizes of aggregates to confirm and detect temporal and spatial distribution patterns of the herbicide during the wind erosion process. Smaller particles can be transported hundreds or even thousands of kilometers, vertically or sporadically horizontally, and these particles are carried far from the starting point (Lyles, 1988), which can cause environmental contamination. In this study, high volumes of both molecules were measured in wind-blown material that is primarily transported by saltation in a horizontal flow movement. Moreover, we should bear in mind that this movement causes the break-up of aggregates, which could become smaller or could, due to their low density, be transported over longer distances. In order to confirm the amount of herbicide transported by suspension over long distances, which potentially can produce environmental contamination, it is necessary to sample particles that are transported by suspension at a greater height, so that in this way we can also measure the concentration of both molecules in PM_{10}fractionated from the sample collected at different heights. Recently, in a laboratory experiment, the fraction PM_{10} contained in different fractions of aggregates have been separated and it was determined that the concentrations of glyphosate and AMPA in those particles are enriched with respect to the source (Méndez et al, 2017).

Our studies are, to our knowledge, the first to measure the concentration of glyphosate and AMPA in the material transported by the wind in field conditions.

According to the results of Méndez et al (2017), the concentrations in the particulate material are higher than those found in the source that is way is probably that glyphosate and AMPA content in the PM10 will be higher than those found in the samples collected in the present investigation. Here, the concentrations of glyphosate and AMPA are recorded in three representative experimental sites that cover an area of 21,410,000 hectares eroded by the wind (FAO, consulted on 03/15/2017). It is important to emphasize the need to monitor to avoid and reduce the drift of pesticides from the plots sprayed on crop production and incorporate soil management practices to preserve the environment.

CONCLUSION

Concentrations of glyphosate and AMPA were found in all soils, including those that never had herbicide application, evidencing the existence of herbicide drift. The soil with the highest agricultural history had the highest concentrations, 131.5µg kg^{-1} glyphosate and 703.5 µg kg^{-1} AMPA.The wind-blown material at a height of 150 cm has average concentrations of 247µg kg^{-1} and 218µg kg^{-1} of glyphosate and AMPA, respectively. The material eroded at 13.5, 50 and 150 cm height was enriched with respect to the original soil at all sites. The concentrations of both molecules found at the CH site, which had never received herbicide, showed the contribution that wind erosion generates from the surrounding regions. The eroded material can, potentially, have a negative impact on the ecosystem.

More studies are needed to determine the amount and concentration of agrochemicals in the material less than 10 µm obtained at different height. Studies are also needed to understand whether such particulate material, which can enter the respiratory system, can affect human health.

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Table 1. Contents of organic matter (OM), analysis of texture, geometrical mean diameter of particles (GDMp) and average diameter of the aggregate (GDMa) of the soils studied.

Location	OM	Clay	Silt	Sand	Sand	Sand	Sand	Textural class	GDMp	GDMa
		< 2 µm	2-50 µm	50 - 74 µm	74-105 µm	105-250 µm	250 - 2000 µm			
	----------	----------	----------	----------	% ----------	----------	----------		µm	µm
Chaco	0.7	6.4	7.0	4.1	10.7	46.9	24.8	Loam Sandy	124	129
La Pampa	2.7	12.9	19.7	12.3	17.6	28.2	9.3	Sandy Loam	63	111
San Luis	1.0	4.9	7.2	4.2	13.7	55.1	14.9	Sandy	119	125

Table 2. Concentration of glyphosate and AMPA in the soil at Los Frentones (Chaco), Santa Rosa (La Pampa) and Villa Mercedes (San Luis).

	CONCENTRATION IN SOIL ($\mu g\ kg^{-1}$)	
Experimental site	Glyphosate	AMPA
Chaco	2,4	6,3
La Pampa	9,5	66,2
San Luis	131,5	703,5

Table 3. Direction of prevailing wind (PD), maximum speed in period of excess (MS), duration of the storm (DS) and horizontal mass transport (HMT) by location and erosion event.

Location / Event Day	PD	MS	DS	HMT
		$m\ s^{-1}$	h min	$kg\ m^{-1}$
CH1 (03/03/2009)	NE	8,8	7h	506,1
CH 2 (29/06/2009)	NE	7,6	41h 25 min	1085,8
CH 3 (31/08/2009)	NE	9,5	28h 30 min	1614,1
CH 4 (07/09/2009)	SW	7,3	40h	772,9
CH 5 (14/10/2009)	NW	9,1	29h 5 min	947,9
CH 6 (04/10/2010)	SE	6,8	72h 30 min	91,8
CH 7 (01/11/10)	SW	8,7	19h 30 min	1104,4
LP 1 (27/12/2004)	wr	wr.		468,6
LP 2 (21/02/2005)	NW	8,6	8h 40 min	644,6
LP 3 (08/04/05)	SE	8,8	2h	986,1
LP 4 (23/10/2009)	SW	12,5	1h 40min	894,4
LP 5 (27/10/2009)	NW	8,4	3h 20min	161,1
LP 6 (16/11/2009)	NE	7,9	8h 30min	772,9
LP 7 (19/11/2009)	SW	10,7	14h 30min	715,1
LP 8 (02/12/2009)	SW	10,7	49 h	2231,4
SL 1 (14/12/2007)	NW	7,9	25h 30min	883,2
SL 2 (18/12/2007)	NW	6,1	7h 45min	14,4
SL 3 (07/01/2008)	wr	Wr		264,8

Reference: wr: without reading, CH: Chaco, LP: La Pampa, SL: San Luis.

Figure 1.Location of experimental sites in Argentina.

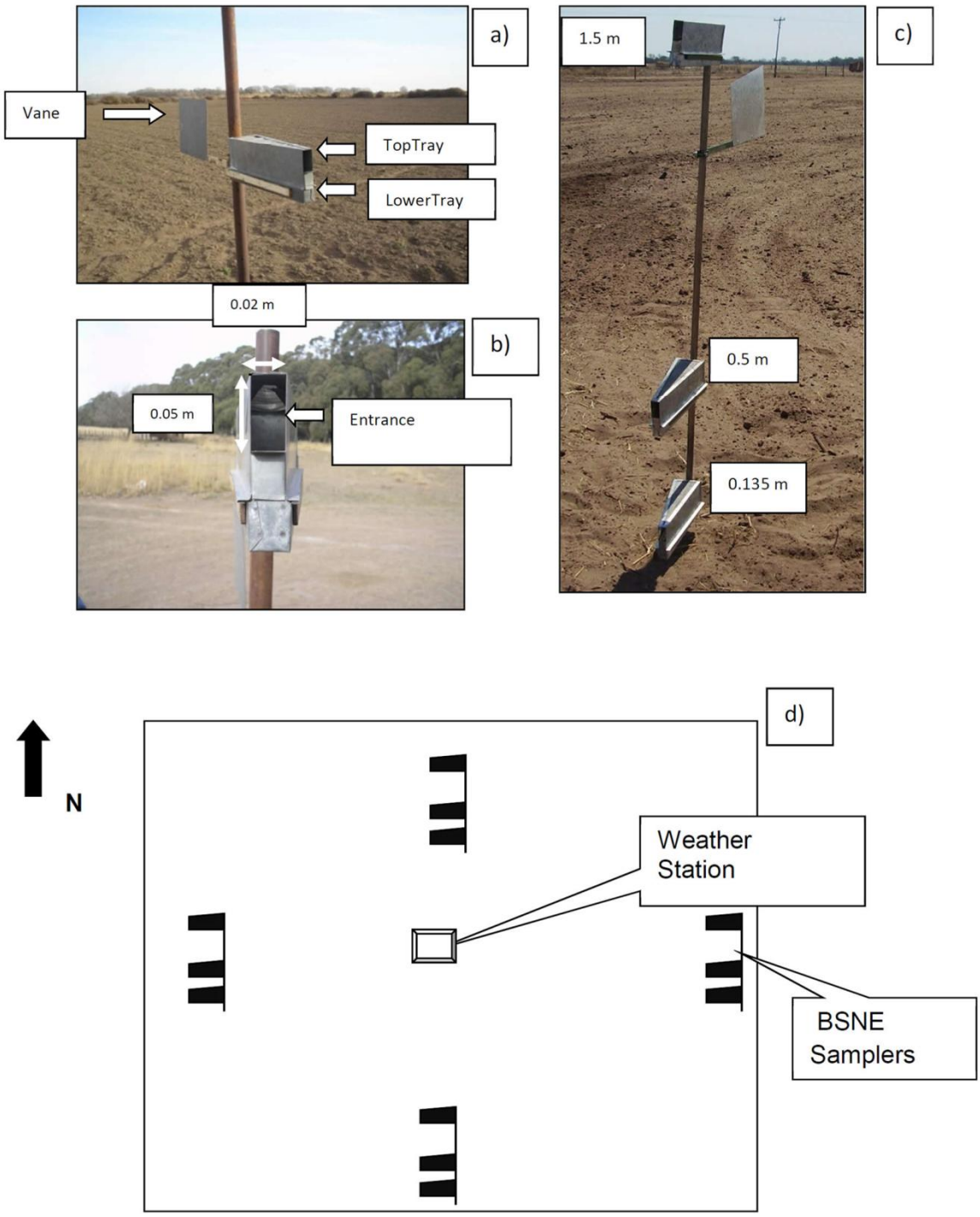

Figure 2.a) Structure of Big Spring Number Eight (BSNE) sampler, b) Details of the opening c) Location of BSNE samplers on each mast d) Location on the plot of masts with samplers and the weather station.

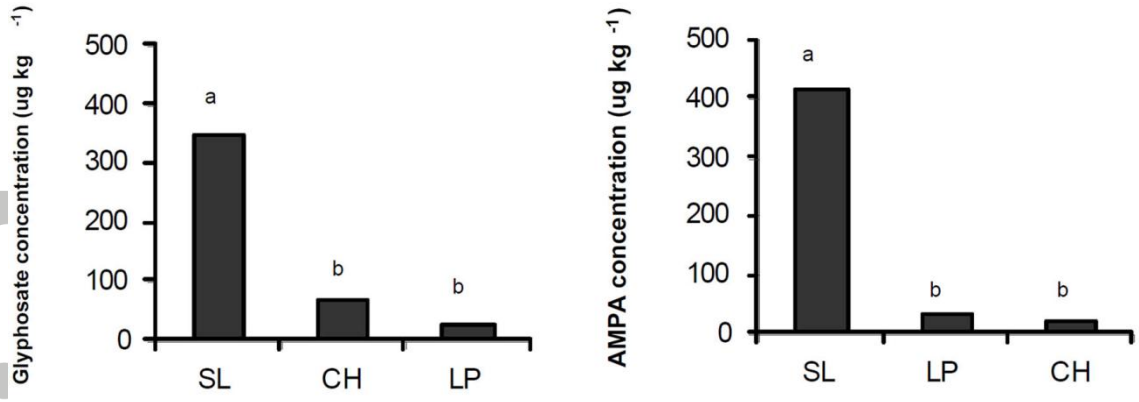

Figure 3. Concentration (µg kg^{-1}) of a) Glyphosate and, b) AMPA in wind-blown materialby location. Different letters indicate statistically significant differences ($p<0.05$).

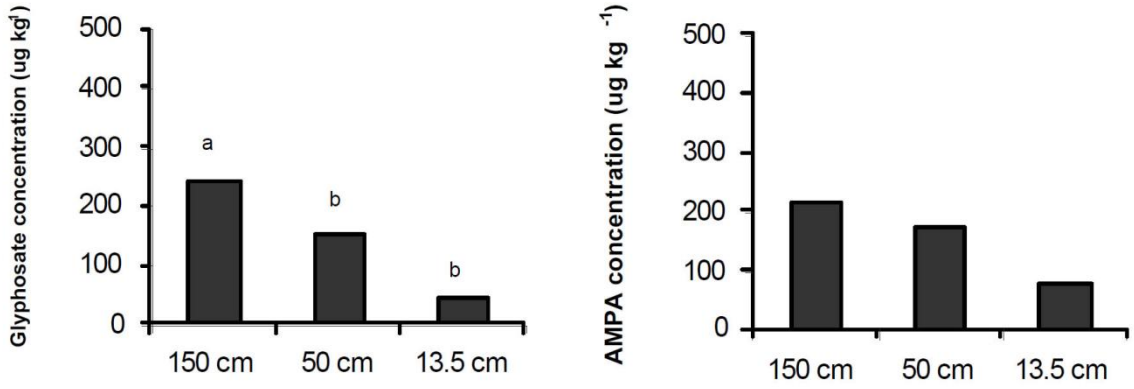

Figure 4.Concentration (µg kg^{-1}) of a) Glyphosate and b) AMPA in wind-blown material by height (cm). Different letters indicate statistically significant differences ($p<0.05$).

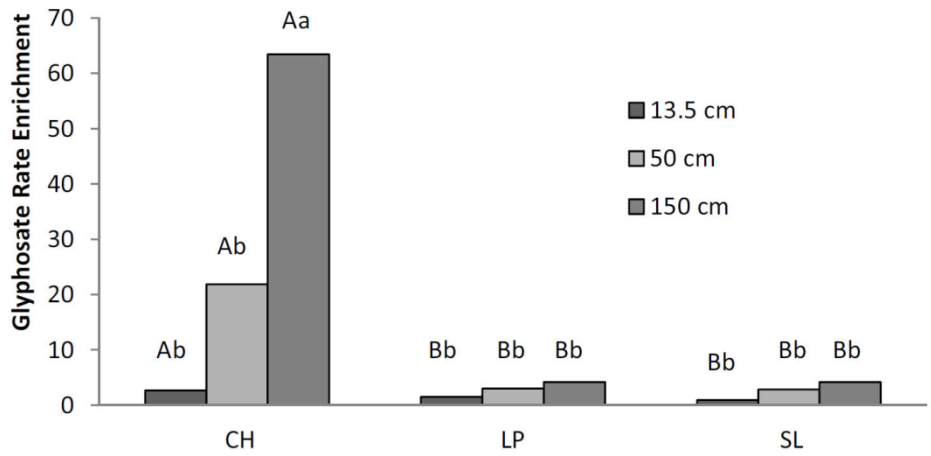

Figure 5.Enrichment rate of glyphosate in the eroded matter at 13.5, 50 and 150 cm at Chaco, La Pampa and San Luis. Different letters indicate statistically significant differences ($p<0.05$): lower-case letters, between height; upper-case letters, between sites.

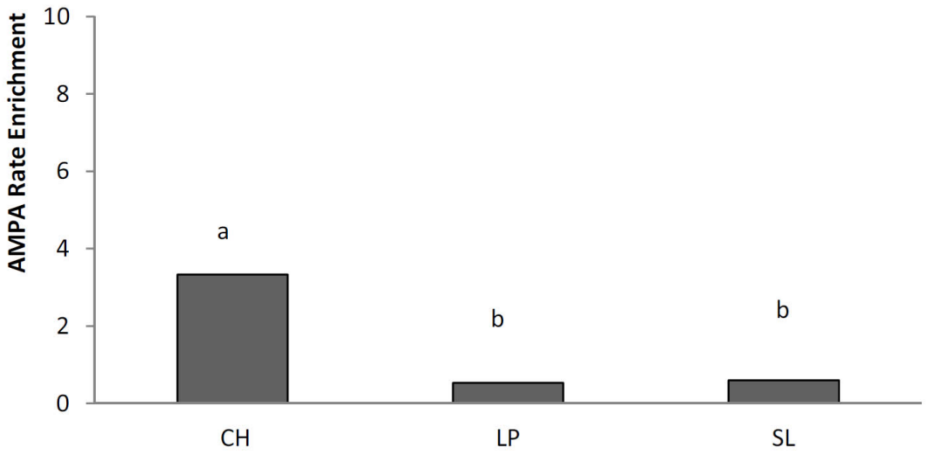

Figure 6.Enrichment rate of AMPA in the eroded matter at Chaco, La Pampa and San Luis. Different letters indicate statistically significant differences ($p<0.05$).